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Review of functional properties of natural fiber-reinforced polymer composites: Thermal insulation, biodegradation, and vibration damping properties

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Natural fiber-reinforced polymer composites are attracting researchers' attention as environmentally friendly and sustainable materials because of their low environmental load imposed by manufacture and disposal. Natural fiber-reinforced composites have comparable or somewhat inferior mechanical performance when compared to glass fiber-reinforced plastics (GFRP), but have some unique characteristics, which include low thermal conductivity, biodegradation in natural environments, and effective suppression of vibration. This paper presents a review of the typical functionalities of natural fiber-reinforced composites including thermal insulation property, biodegradable property, and vibration damping property. As described herein, the author will introduce each functionality and discuss the effects of many factors affecting the performance of each function. Consequently, this paper reports that composite materials that fully use their functionality rather than mechanical performance is important for the future development of natural fiber-reinforced composites.

Keywords: biodegradability, functionality, green composites, natural fiber composites, thermal insulation, vibration damping

1. Introduction

In response to growing public awareness of global environmental difficulties, efforts to realize a sustainable and recycling-oriented society are now progressing in various fields [1]. Given these circumstances, it has been recognized that reducing the environmental loads of glass fiber-reinforced plastics (GFRP) and carbon fiber-reinforced plastics (CFRP), which are being

used in large quantities as lightweight structural materials, is an important environmental issue in the composite materials industry [2]. For this reason, research and development has been conducted on such subjects as the recovery of glass fibers from GFRP wastes through thermal decomposition [3, 4]. In fact, our society has been requesting the development of environmentally friendly composite materials having mechanical characteristics equivalent to those of GFRP [5, 6, 7, 8, 9, 10, 11].

In the polymer industry, mass production of polylactic acid (PLA), among the most commercialized biodegradable resins, began in the 1990s [12]. Responding to this new trend in the polymer industry, extensive research has been conducted on environment-friendly composite materials having a combination of natural fibers and a biodegradable resin: so-called green composites [13, 14, 15, 16, 17, 18, 19, 20, 21]. In the early stage of this research in this field, the main purpose of many research efforts was to improve the mechanical properties of green composites such as strength and the elastic modulus [22, 23, 24, 25, 26, 27, 28, 29, 30, 31]. Among the wide variety of efforts, surface modification of natural fibers [32, 33, 34] and hybridization of the reinforcing natural fiber [35, 36, 37] were initiated to increase green composites' mechanical performance. From that enthusiastic research and development effort, some natural fiber reinforced composites have been applied to some commercial equipment [38, 39, 40, 41].

It is noteworthy that green composites have not only a low environmental load (their carbon footprint is small) but also a unique function of biodegradability [42, 43, 44, 45, 46],

which no conventional composite material has. In addition to this biodegradability function, natural fiber-reinforced composites have other unique functionalities such as heat-insulating properties [47, 48, 49, 50, 51, 52, 53] and vibration damping properties [40, 54, 55, 56, 57, 58, 59].

Many review papers describing the mechanical or chemical characteristics of natural fiber-reinforced composite materials have been published to date [8, 9, 16, 60, 61, 62, 63, 64, 65, 66]. However, limited information is available on the functionality of natural fiber-reinforced composites. Therefore, the scope of this paper is a review of the typical functionalities of natural fiber-reinforced composites including thermal insulation, biodegradability, and vibration damping.

2. Origin of functionalities of natural fibers

The internal microstructure of a typical natural plant fiber is portrayed in Figure 1 [67]. Macroscopic one plant natural fibers consist of many elementary fibers [68, 69]. Each elementary fiber has a cavity. Therefore, a hole can be found in the cross-section of the elementary fiber. This air-filled cavity is called a lumen. Most natural fibers are light. In addition to providing this characteristic of light weight, more importantly, this air-filled lumen plays a role in improving heat retention and also hygroscopic/moisture releasing properties of the natural fiber [70]. This function of natural fibers is still valid in a polymer composite material. Consequently, some functionalities of natural fiber-reinforced composites are attributable to this lumen.

The lumen size and morphology are known to be changed remarkably depending on the fiber species, as shown in Figure 2 [69]. Furthermore, the abaca fiber lumen size can be altered by chemical treatment, as reported earlier by Liu et al. [71] and Cai et al. [72]. It is noteworthy from these results that the internal structural changes brought about by the chemical treatment can control the natural fiber-reinforced composites' functionality.

Three main constituents of the natural plant fiber are cellulose, hemicellulose, and lignin [70, 73]. The cellulose and hemicellulose are hydrophilic, but the last one, lignin, has hydrophobicity. Therefore, the first two constituents play an important role in functionalities related to the chemical reactions with water or vapor, such as biodegradability or moisture absorption and/or desorption [74, 75].

3. Thermal insulation properties

Some natural fibers such as cotton and flax have been used as raw materials for clothing because they have a soft feeling and have a large lumen in the fiber. Therefore, their heat insulation property is better than those of other natural fibers such as jute and hemp. With that knowledge, the thermal properties of natural fibers can be predicted to differ greatly from those of glass fibers.

Agrawal et al. [47] examined the thermal properties of oil palm fiber-reinforced phenol formaldehyde (PF) composites. They reported the effects of three surface treatments on the thermal conductivity and thermal diffusivity of the composites with 40wt.% oil palm fiber: silane treatment, alkali treatment, and acetylated treatment. Results show that both the thermal conductivity and thermal diffusivity of silane-treated or alkali-treated fiber were higher than those

of untreated fiber. Therefore, both thermal conductivity and thermal diffusivity of the composites increased after surface treatment. However, they did not report the effects of fiber content on the thermal conductivity of the composites.

Mangal et al. [48] examined the effects of pineapple leaf fiber content on the thermal conductivity of the PF-based composites. They reported that the thermal conductivity of the composites was lower than that of neat PF resin (the thermal conductivity of the composites with 46.4 vol.% fiber was 34% lower than that of neat PF resin) and that the thermal conductivity of the composites decreased concomitantly with increasing fiber volume content. They stated that the reason for the lowering of the thermal conductivity of the composites was that the pineapple leaf fiber had lower thermal conductivity than that of PF resin.

Singh et al. evaluated the temperature dependence of the thermal conductivity of a similar composite system, oil palm fiber-reinforced PF composite, at temperatures of 50°C to 110°C [49]. They also examined the effect of the three surface treatments (silane, alkali, and acetylate) on the temperature dependence of the thermal conductivity of the composites reinforced by 40 vol.% of oil palm fiber. They pointed out that a clear peak in thermal conductivity was observed at around 90°C for each surface-treated composite. They demonstrated that this peak temperature was close to the glass transition temperature of the composite system. Furthermore, they proposed that this peak is attributable to microstructural changes in the polymer matrix, specifically the crosslinking density of the matrix resin became minimal, and that it produces a maximum value of the phonon

mean free path. A similar temperature dependence related to the glass transition point has also been reported for poly-L-lactic acid (PLLA) [76].

The sample density effects on the thermal conductivity of fully biodegradable bamboo fiber-reinforced PLA based green composites were evaluated both experimentally and theoretically [50]. Flocculent bamboo fibers of about 20 μm diameter were extracted from steam-exploded bamboo columns. Bamboo fiber-reinforced green composites (BFGC, bamboo fiber contest = 60 wt.%) with different densities were prepared by adjusting the molding conditions, such as the molding pressure and molding time. Reportedly, the density dependence of the thermal conductivity of BFGC was well described using Russel's model [77]. The thermal conductivity of the BFGC increased as the sample density increased, as presented in Figure 3. They also described that the heat insulation properties of the BFGC were comparable to those of natural woods and that they were better than those of GFRP and CFRP (Figure 4).

Liu et al. [51] first reported the importance of the lumen size of the reinforcing natural fiber for the interpretation of thermal conductivity in an abaca fiber-reinforced composite system. They explained, based on results of theoretical and numerical analyses, the effects of the size and distribution of the lumen in the single natural fiber on the transverse thermal conductivity of a simple unit cell model with a single fiber. Important points to note are that the transverse thermal conductivity of single fiber composite system depends only on the volume fraction of the lumens, and that the thermal conductivity decreases with an increasing volume fraction of the lumens. However, it is independent of their shape and distribution.

Liu et al. [71, 78, 79], using the results described above, reported the effects of geometrical parameters (fiber volume fraction and lumen size) and thermal properties of the constituent materials (thermal conductivity values of both matrix resin and fiber) on the transverse thermal conductivity of the composite materials. They found that the dependence of the transverse thermal conductivity on the fiber volume fraction varies with the fiber lumen size and the thermal conductivity values of the constituent matrix and fiber. It is noteworthy that the dependence of the transverse thermal conductivity on the fiber content is reversed by the lumen size of the fiber, as presented in Figure 5 [78]. For instance, for composites reinforced by a fiber with a small lumen (small α), the transverse thermal conductivity increases as the fiber content increases, but in the case of the composites reinforced by the fiber with large lumen (large α), the transverse thermal conductivity decreases. This unique dependence on fiber content was confirmed experimentally using bamboo (small lumen) and abaca (large lumen) fibers [69]. Similar dependence related to the lumen size effect was reported elsewhere [80, 81].

The lumen size in the natural fiber used as a reinforcing phase is the most important factor to obtain composites with lower thermal conductivity, as described above. However, the situation is completely different in nanocellulose fiber (NCF) reinforced composites, for which there is no lumen in the NCF. Shimazaki et al. [82] have reported high in-plane thermal conductivity of 1.1 $Wm^{-1}K^{-1}$ in epoxy-58wt.%NCF composites. They also pointed out that the NCFs can transport more phonons through the epoxy-NCF composites, indicating that the NCFs are responsible for the marked improvement in the thermal conductivity of the composite. Similar improvement in

thermal conductivity in NCF-reinforced polymer composites has been reported elsewhere [83, 84].

4. Biodegradable properties

A characteristic and important functionality of green composites is biodegradability, which derives mostly from the action of microorganisms [60], even for natural fiber-reinforced composites with a non-biodegradable matrix. Some examples are flax fiber-reinforced polypropylene composites, which are able to biodegrade in an appropriate environment [85]. As results of many studies have indicated, biodegradability is an effective and indispensable function for the waste treatment of green composites after completing their service life. However, the introduction of biodegradability in composites simultaneously brings about lower durability. Therefore, control of biodegradability behavior is needed. Ideally, a trigger function for starting the biodegradation reaction of the green composites is desired.

Several studies have examined biodegradability for various biodegradable plastics such as starch, polyvinyl alcohol (PVA), PLA, and polyhydroxybutyrate (PHB) [86]. Because of its unique bioabsorbable nature, the biodegradable polymer PVA was applied to suture threads in the early 1930s [87]. After this step, research on biodegradable composite materials began in the same medical field [88], but not in the field of engineering. Vainionpää et al. [89] developed self-reinforced, bioabsorbable polyglycolic acid (PGA) composite rods for fracture fixation instead of conventional metallic rods. The self-reinforced polymer composites consist of a polymer matrix with reinforcing fibers of the same polymer. Vainionpää et al. reported that the

self-reinforced PGA composite rods had sufficient flexural strength of 250 MPa. Törmälä et al. [90] found that the PGA/PLA fiber self-reinforced composites also had sufficient mechanical properties for application in the treatment of fractures in the cancellous bone. Törmälä et al. [90] and Alexander et al. [91] also fabricated carbon fiber-reinforced PLA composites. Reportedly, the strength of the composites decreases in about 4 weeks because of the biodegradation reaction, even in the case of PLA-based composites reinforced with carbon fiber having no biodegradability. However, its biodegradability mechanism was not described. It is noteworthy that, in the case of biodegradable resin matrix composites, the composites can be biodegraded irrespective of the biodegradability of the reinforcing fiber.

Belmares et al. [92] fabricated natural fiber-reinforced thermoset composites (fiber loading = 30–35 wt.%) and evaluated their biodegradation properties compared with those of glass-fiber reinforced thermoset composites (fiber loading = 51–55 wt.%). A natural fiber mat (450 g/m^2) made with 90-mm-long randomly oriented palm fiber was used as reinforcement in natural fiber-reinforced composites. For glass fiber-reinforced composites, a woven glass mat (500 g/m^2) was used. Their long term biodegradation behavior for 300 days was monitored using a soil burial method. The degree of biodegradation was evaluated according to the change in tensile strength. For glass fiber-reinforced composites, the strength was almost constant, although the strength of natural fiber-reinforced composites decreased concomitantly with increasing burial time, indicating that the natural fiber-reinforced thermoset composites can be biodegraded in the natural environment [93].

Akahori and Osawa [42] reported the biodegradation behavior of wood paper/polycaprolactone (PCL) biodegradable composites during soil burial. They described that the PCL-laminated paper composites crumbled within two months. The composites were biodegraded faster than the neat PCL. Both the number average molecular weight and weight average molecular weight of the residual PCL samples decreased gradually during soil burial experiments.

Shibata et al. [43] fabricated three green composites with polybutylene succinate (PBS), polyestercarbonate (PEC)/PLA blend, and PLA as matrix polymers by injection molding. The green composites were reinforced by short abaca fibers (about 5 mm) that were surface-treated by esterification with acetic anhydride (AA-abaca), butyric anhydride treatment (BA-abaca), alkali treatment (Alk abaca), and cyanoethylation (AN abaca). The biodegradability of the neat matrix polymer and the green composites reinforced by AA-abaca and untreated abaca fibers was measured by evaluating the weight loss of the film samples buried in soil (1:1 mixture of leaf mold and black soil for gardening) for 24 weeks, as shown in Figure 6. The noteworthy point is that the biodegradability of PLA resin that is only slightly biodegraded in the soil can be improved greatly by compositing with non-treated abaca fiber.

Another important point is that the biodegradation behavior of the green composites reinforced by acetylated abaca fiber resembles that of neat resin irrespective of the type of matrix polymer. Moreover, the acetylation treatment on the abaca fiber can suppress the biodegradability of the green composites [43]. They concluded that the green composites reinforced by untreated

abaca fiber have greater loss than either the neat matrix resin or the green composites reinforced with AA-abaca fiber (acetylated fiber). Similar enhanced biodegradation behavior was also reported for natural or cellulosic fiber-reinforced composites [44, 94].

According to the literature [45], the biodegradation characteristics of both abaca fiber and unidirectional abaca fiber reinforced starch-based green composites (fiber content = 50 wt.%) were examined using a household garbage disposal. The changes in the tensile strength of abaca fiber and composite (hereinafter biodegradation treatment) were measured. The strength of abaca fibers decreased sharply after 2–5 days of treatment. Thereafter, the strength was lowered gradually (Figure 7(a)). Subsequently, the abaca fibers were biodegraded and divided into short fiber pieces after 20 days of biodegradation. They were partially biodegraded into fine elementary fibers of about 15 μm diameter. Reportedly, the tensile strength of abaca fiber, which had high strength of 748 MPa before the biodegradation test, decreased to about 17% (124 MPa) after biodegradation treatment of 15 days. However, the tensile strength of the starch-based green composites decreased sharply after two days of biodegradation treatment. Thereafter, the strength tended to decrease gradually after 15 days (Figure 7(b)). This dependence is the same as the strength change of abaca fiber described above. This finding suggests that the strength change attributable to the biodegradation treatment of the green composite is caused mainly by the change in the reinforcing fiber strength.

Ochi et al. [45] proposed a biodegradation sequence (Figure 8) for the green composites based on microscopic observation of morphological features of biodegraded sample surfaces.

First, the biodegradable resin at the specimen surface decomposes; then abaca fiber bundles embedded in the starch-based resin are exposed. Next, the biodegradable resin near the interface between abaca fibers and the biodegradable resin decomposes, which causes the formation of an interfacial gap. Finally, the decomposition of both abaca fiber bundles and biodegradable resin occurred remarkably.

The next important factor affecting the biodegradable characteristics of green composites is water absorption. As reported by Carvalho et al. [70], water absorption in green composites increased with fiber loading. The composites showed faster water absorption than that which occurred near matrix resin alone. Therefore, the matrix resin and reinforcing natural fibers play an important role, just as the interface does, in the biodegradation behavior of green composites.

5. Vibration damping properties

Vibration is sometimes a desirable physical phenomenon. However, successive vibration in structural materials often causes difficulties such as generation of noise and introduction of fatigue cracks [95]. Therefore, the development of materials with better vibration damping properties is becoming increasingly important year-by-year.

Another unique and effective function of green composites is their vibration damping [96, 97, 98, 99, 100, 101]. Natural fibers are known to have more viscoelastic properties than inorganic fibers such as glass and carbon fibers [102]. It is possible that polymer composites with better vibration damping properties are obtainable using natural fibers as reinforcements [103, 104, 105]. As with other GFRP and CFRP, the damping characteristics of green composites are

dominated by many factors such as fiber surface treatment, fiber loading, fiber length, and architecture of the reinforcing fiber. Details of the effects of these factors are described below.

Saha et al. [96] used dynamic mechanical analysis (DMA) to study the dynamic mechanical properties of surface-treated jute-fiber-reinforced unsaturated polyester composites at temperatures of 40°C to 190°C. The results demonstrated that the loss factor (tan δ) peak value of the composites was lower than that of neat resin, as shown in Figure 9. This phenomenon has also been reported for other systems [106]. Apparently, the incorporation of stiffer fibers decreases the tan δ peak value by restricting the molecular movement of the matrix polymer. However, the tan δ values of the composites reinforced by non-treated jute fiber at lower temperatures from 40°C to 80°C (around the peak temperature of the composites) were higher than that of neat resin. This result indicates that the incorporation of natural fiber into a thermoset resin contributes to the improvement of vibration damping properties.

Flynn et al. [107] used a single cantilever vibration test to compare the vibration damping performance of carbon and flax fiber-reinforced composites. They reported that the flax fibers have greater damping properties than carbon fibers have (Figure 10). Moreover, they explained that these superior damping characteristics in flax fiber composites are derived from the higher shock absorbing capability of flax fibers: the elementary flax fibers act as tiny shock-absorbers in the composite system. A similar result has been reported for luffa cylindrica fiber- [108] and flax fiber- [101] reinforced epoxy composite systems. Damping factors of the natural fiber-reinforced composites were higher than those of glass fiber-reinforced composites.

Another factor affecting the damping properties of natural fiber composites is their water content [35, 109, 110]. Cheour et al. [110] reported the effects of water aging on the damping properties of quasi-unidirectional flax fiber-reinforced epoxy composites. They demonstrated that the water uptake causes a decrease in the bending modulus and an increase in the loss factor, as shown in Figure 11. They explained that the reason for this dependence lies in increasing energy dissipation at the interface between resin matrix and flax fiber derived from water absorption. Similar dependence was also found earlier for GFRP [111] and CFRP [112, 113].

The author's research group [114] has examined the vibration damping properties of two types of bamboo fiber reinforced starch-based green composites: randomly oriented short bamboo fiber composites and unidirectional long bamboo fiber composites. Results demonstrated that longer bamboo fibers of the green composites have lower damping factors. This fiber length dependence is not limited to natural fiber-reinforced composites. In fact, Subramanian et al. [115] reported similar dependence on fiber length in glass fiber-reinforced polypropylene composites. Rezaei et al. [116] also described a decrease in the loss factor in carbon fiber reinforced polypropylene composites with increased fiber length. Another important result is that the loss factor of the composites decreased concomitantly with increasing fiber content without dependence on the fiber length. Similar fiber content dependence was also reported in cellulose-fiber-filled polyacetal composites [58] and in glass/ramie polymer composites [117]. Such dependence of damping performance on fiber length and fiber contents are interpreted as being caused by restriction of matrix polymer molecules near the rigid fibers [115].

Because natural fiber reinforced composites have excellent vibration damping characteristics as described above, several interesting applications have been reported. Some researchers have tried applying a natural fiber-reinforced composite panel to the top plates of string musical instruments such as violins, ukuleles, and guitars [118, 119, 120]. Another unique application is as structural members in racing bicycles because flax/carbon hybrid composites have a suitable combination of mechanical and damping performance [121].

6. Conclusions

The present work has explained that natural fiber-reinforced polymer composites not only have acceptable mechanical performance; they also have many unique functional properties such as thermal insulation, biodegradability, and vibration damping capacity. The source of such unique functionalities derives from both the chemical properties and microstructural features of natural plant fibers. It is noteworthy, however, that the degree of the function to be developed varies depending on the type of natural fiber that is used. Therefore, the selection of natural fiber used as reinforcement is an important issue to achieve acceptable functional performance. Important points related to the functionality of the natural fiber-reinforced composites are summarized as explained below.

(1) The thermal conductivity of the natural fiber-reinforced composites is lower than that of GFRP and CFRP with similar fiber contents. The thermal properties of the natural fiber-reinforced composites vary depending on the internal microstructure of the natural fiber, which is determined mainly by the lumen size. In the case of composites reinforced by natural

fibers with a large lumen, the thermal conductivity decreases concomitantly with increasing fiber content. This dependence of thermal conductivity on the fiber content is opposite to that of conventional GFRP and CFRP.

(2) Green composites can be fully biodegraded under composting conditions. Even for natural fiber-reinforced non-biodegradable synthetic polymer composites, biodegradation occurs. The weight loss of the composites can be detected. The natural fiber and the interface play important roles in the biodegradation behavior of the natural fiber-reinforced composites.

(3) Natural fiber reinforced composites have a higher loss factor than conventional GFRP and CFRP. Similarly to the GFRP, longer reinforcing natural fibers lead to lower damping factors of the composites. The loss factor of a composite decreases concomitantly with increasing fiber content. Therefore, hybridization with other natural or synthetic fibers is expected to be important to obtain integrated performance such as high strength and high vibration damping capacity.

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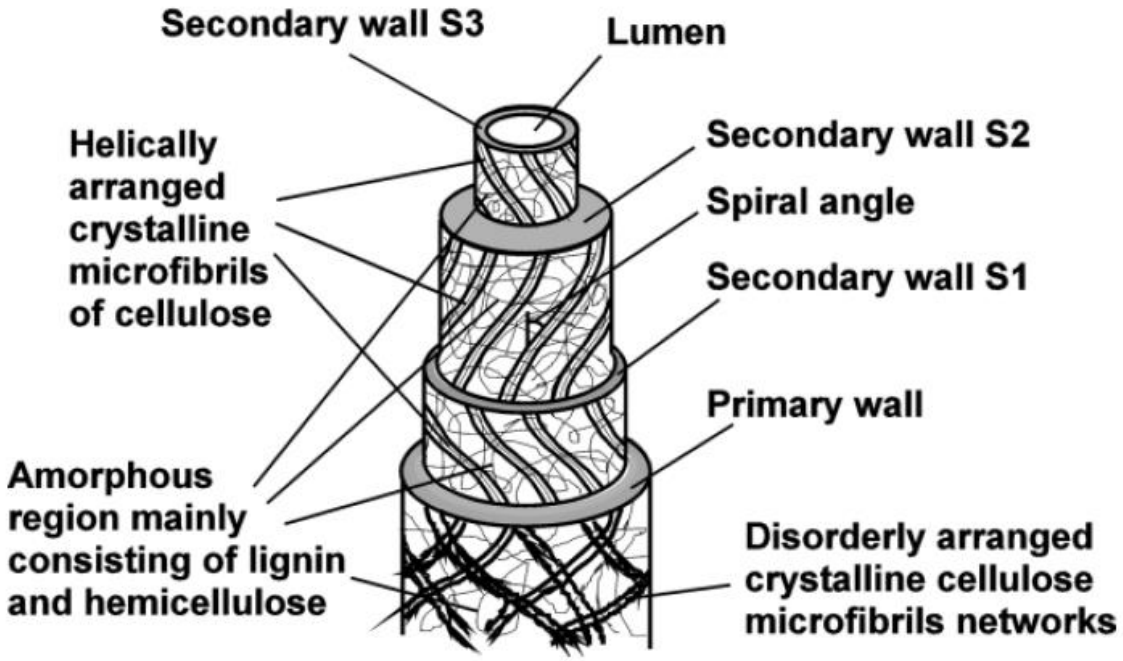

Figure 1. Internal microstructure of natural plant fiber. Reprinted with permission from Elsevier [67].

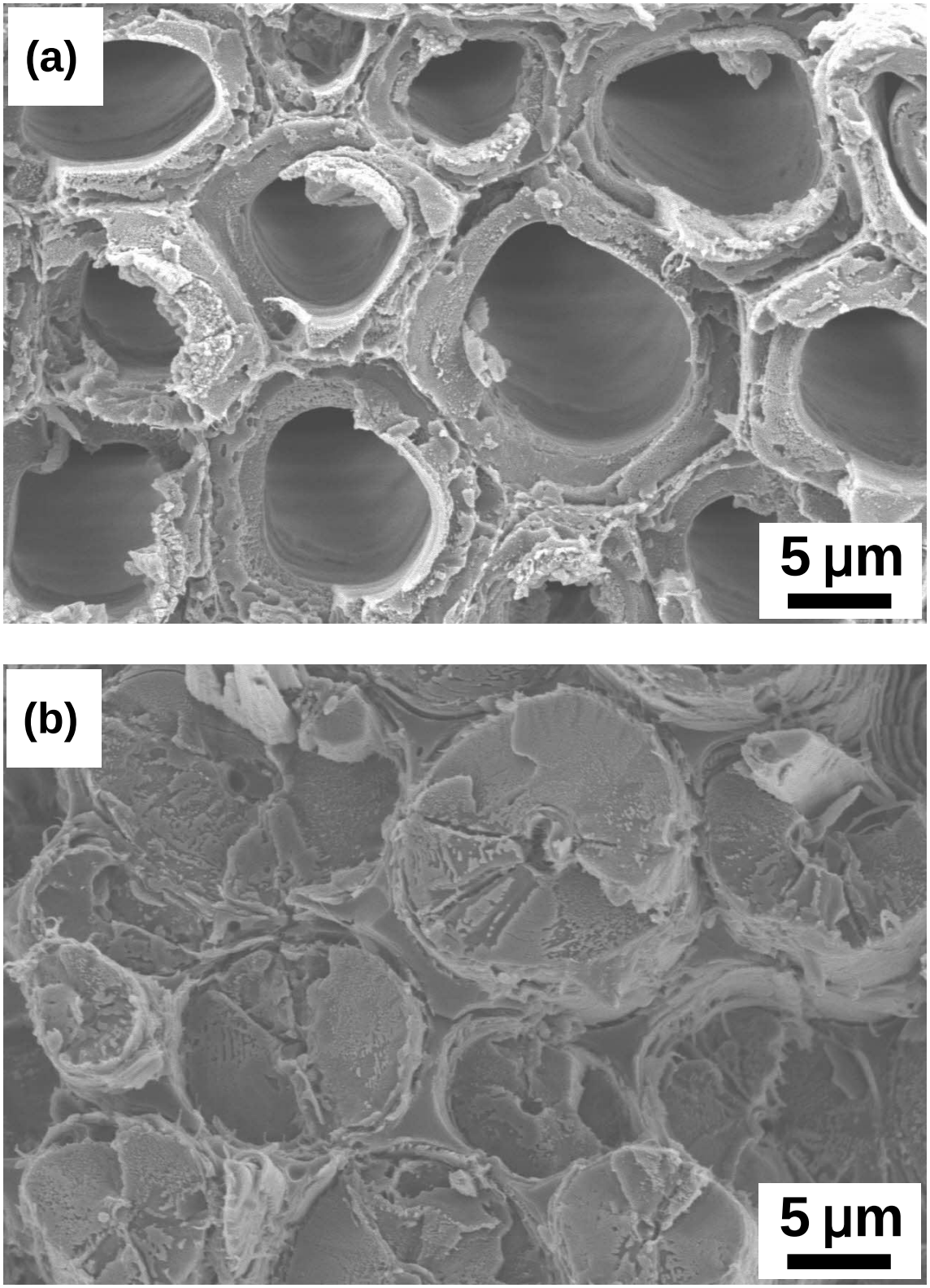

Figure 2. Photomicrographs of internal microstructure of (a) abaca fiber and (b) bamboo fiber, showing the difference in lumen size. Reprinted with permission from Elsevier [69].

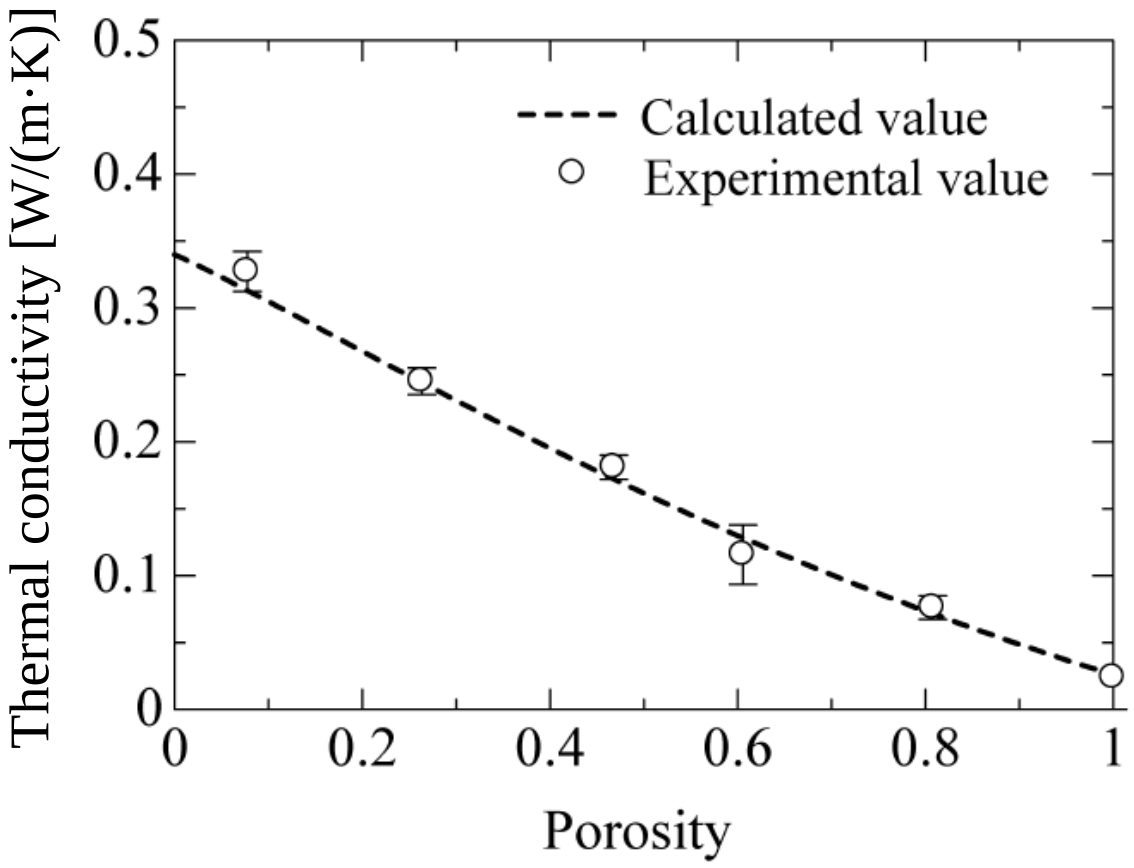

Figure 3. Relation between thermal conductivity and porosity of BFGC. Reprinted with permission from Taylor & Francis [50].

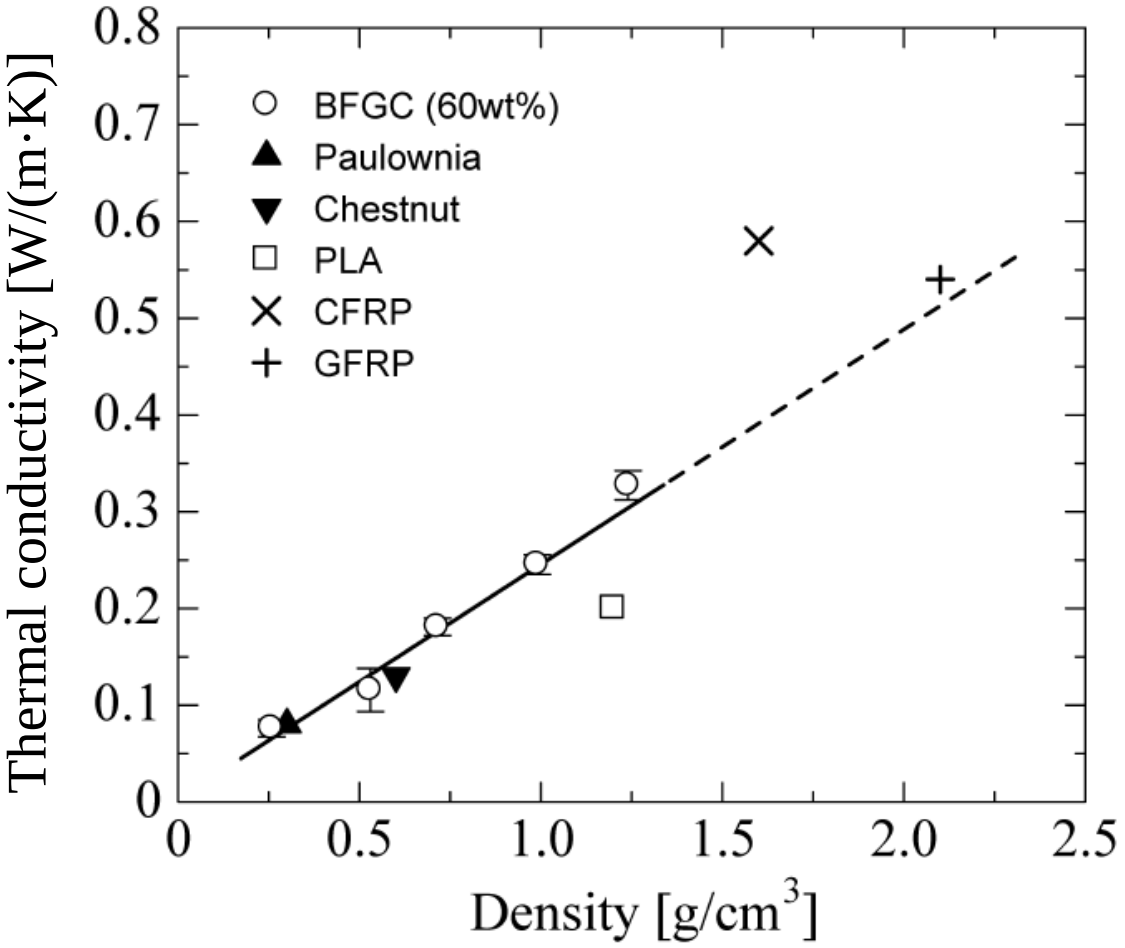

Figure 4. Relation between thermal conductivity and density of BFGC, woods, PLA, and FRPs. Reprinted with permission from Elsevier. Reprinted with permission from Taylor & Francis [50].

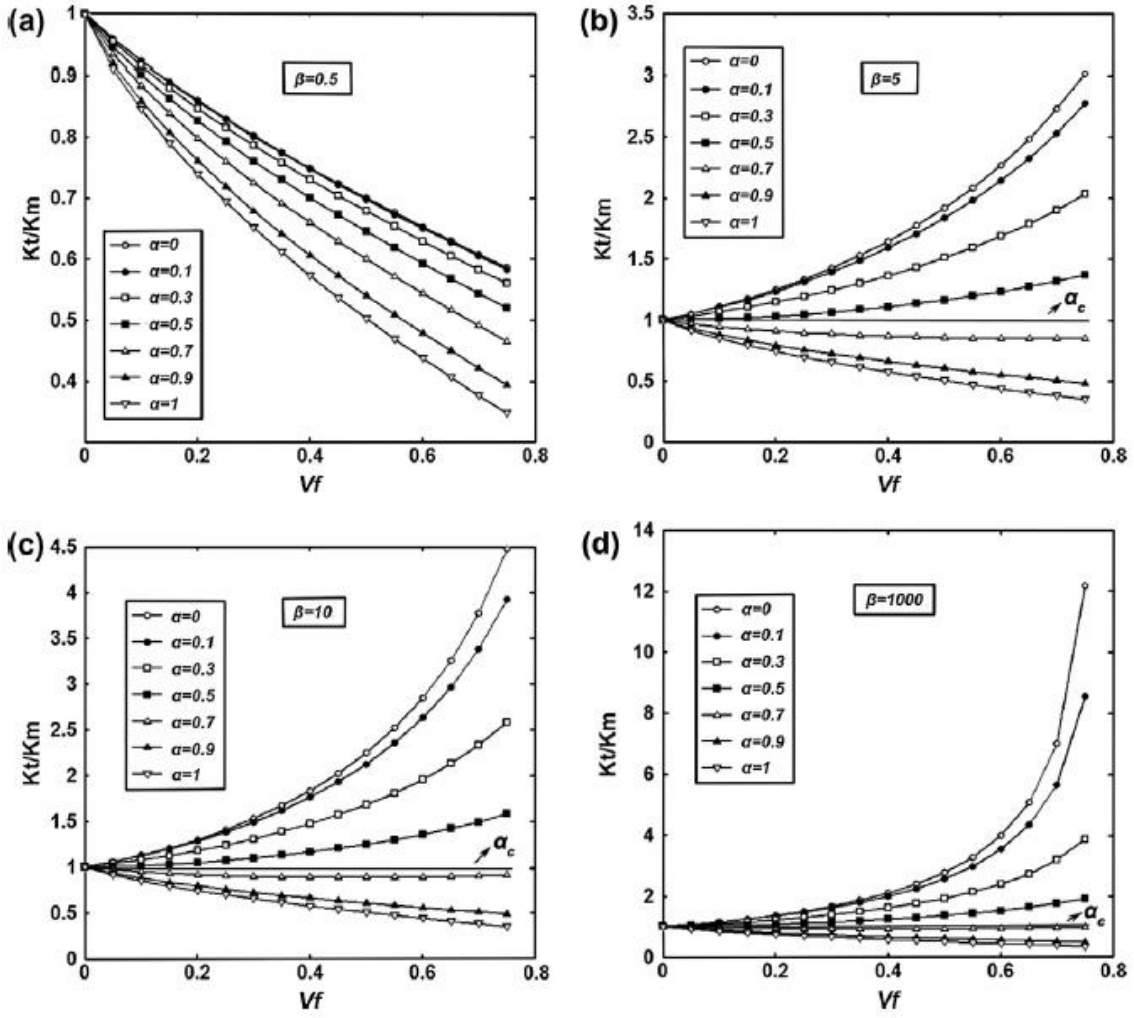

Figure 5. Relation between normalized thermal conductivity of model composite (K_t/K_m) and fiber volume content (V_f) of unidirectional hollow fiber composites as a function of normalized lumen size (α = lumen's radius/fiber's radius) when (a) $\beta = 0.5$, (b) $\beta = 5$, (c) $\beta = 10$, and (d) $\beta = 1000$, where K_t, K_m, and K_f respectively represent thermal conductivity of the model composite, matrix resin, and solid part of fiber, and β is the normalized thermal conductivity of the solid part of the fiber ($\beta = K_f/K_m$). Reprinted with permission from Elsevier [78].

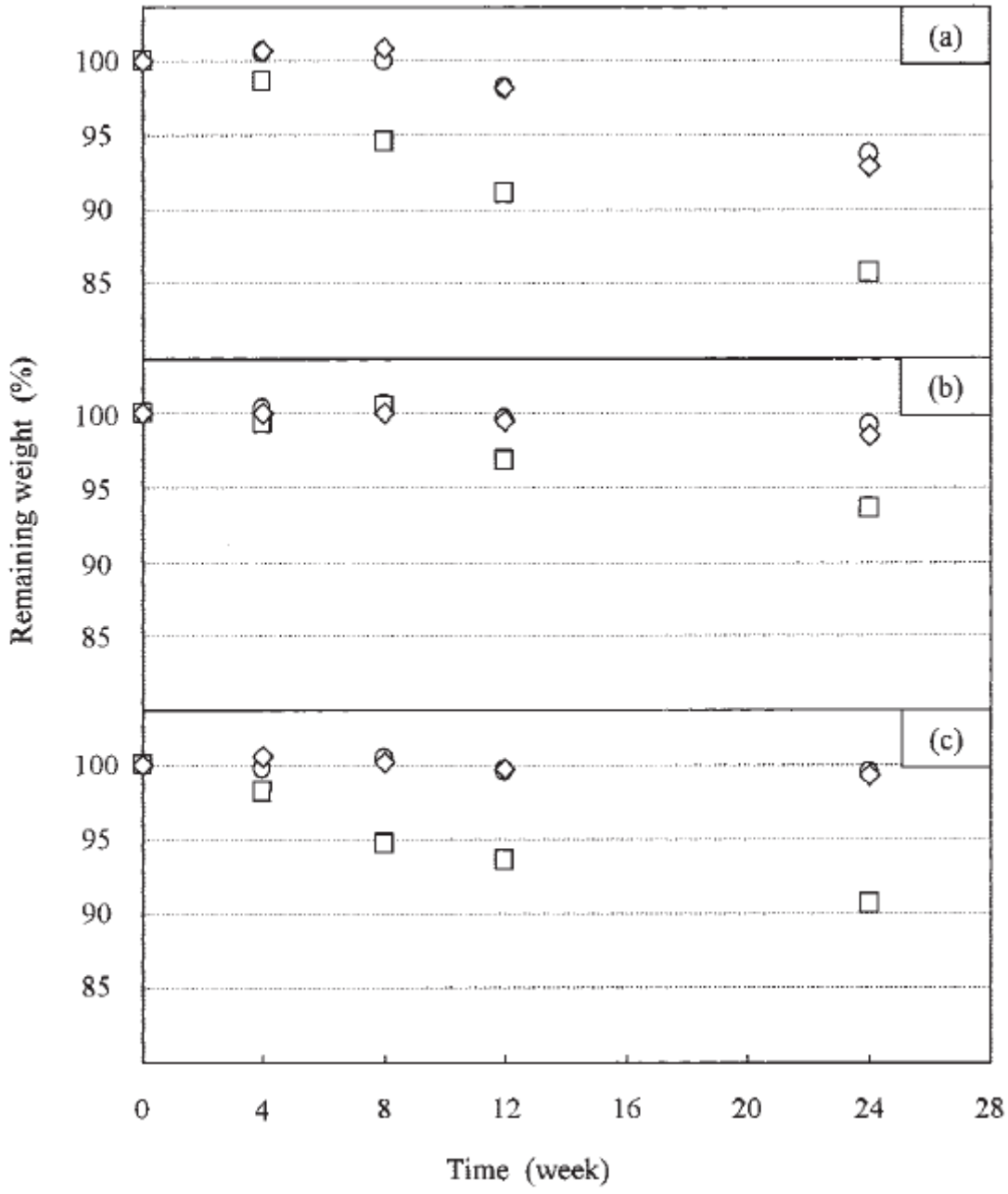

Figure 6. Biodegradation behavior (change in weight loss) of neat resin and composites in a soil-burial test (○, neat resin; □, composites with 10 wt.% untreated abaca fiber; ◇, the composites with 10 wt.%AA-abaca fiber): (a) PBS and PBS composites, (b) PEC/PLA and PEC/PLA composites, and (c) PLA and PLA composites. Reprinted with permission from John Wiley and Sons [43].

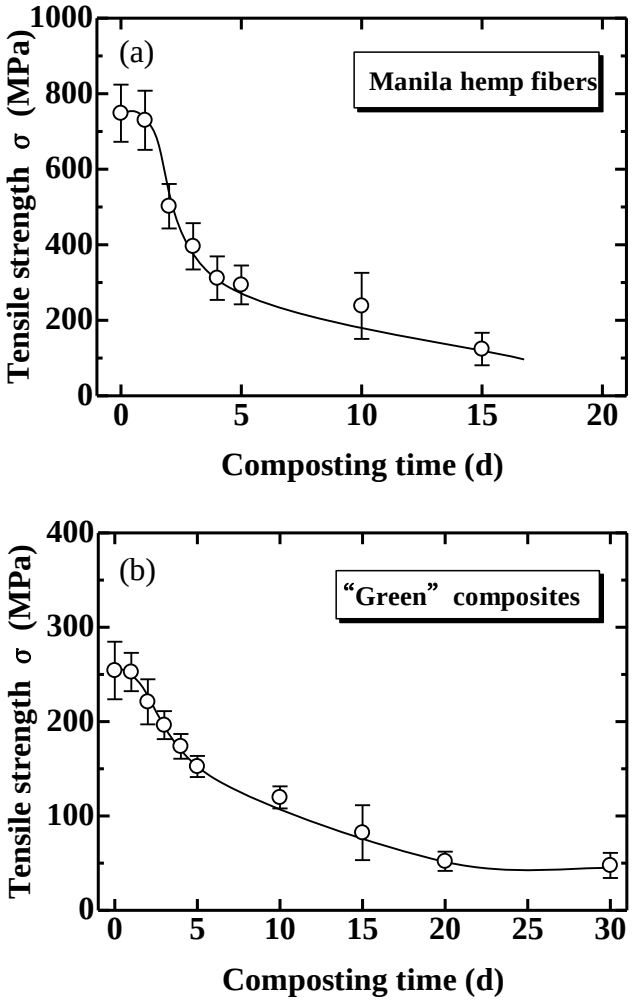

Figure 7. Variation in tensile strength of (a) abaca fiber (Manila hemp fiber) and (b) unidirectional abaca fiber-reinforced green composites. Reprinted with permission from The Society of Materials Science, Japan [45].

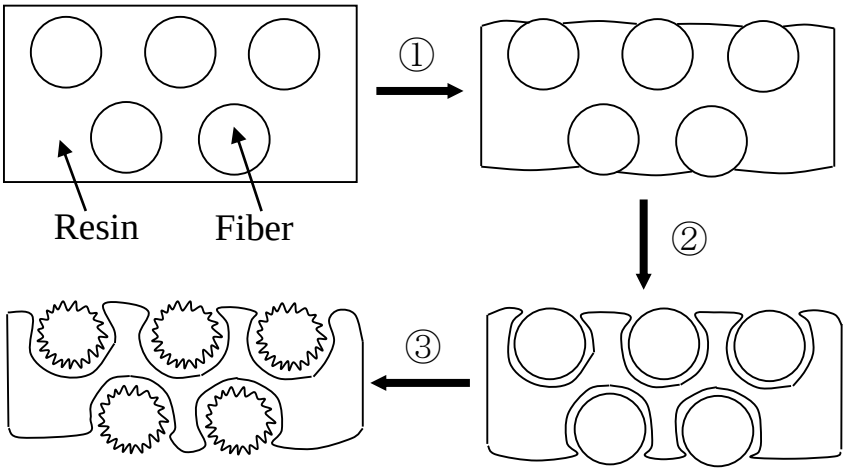

Figure 8. Biodegradation sequences in green composites. Reprinted with permission from The Society of Materials Science, Japan [45].

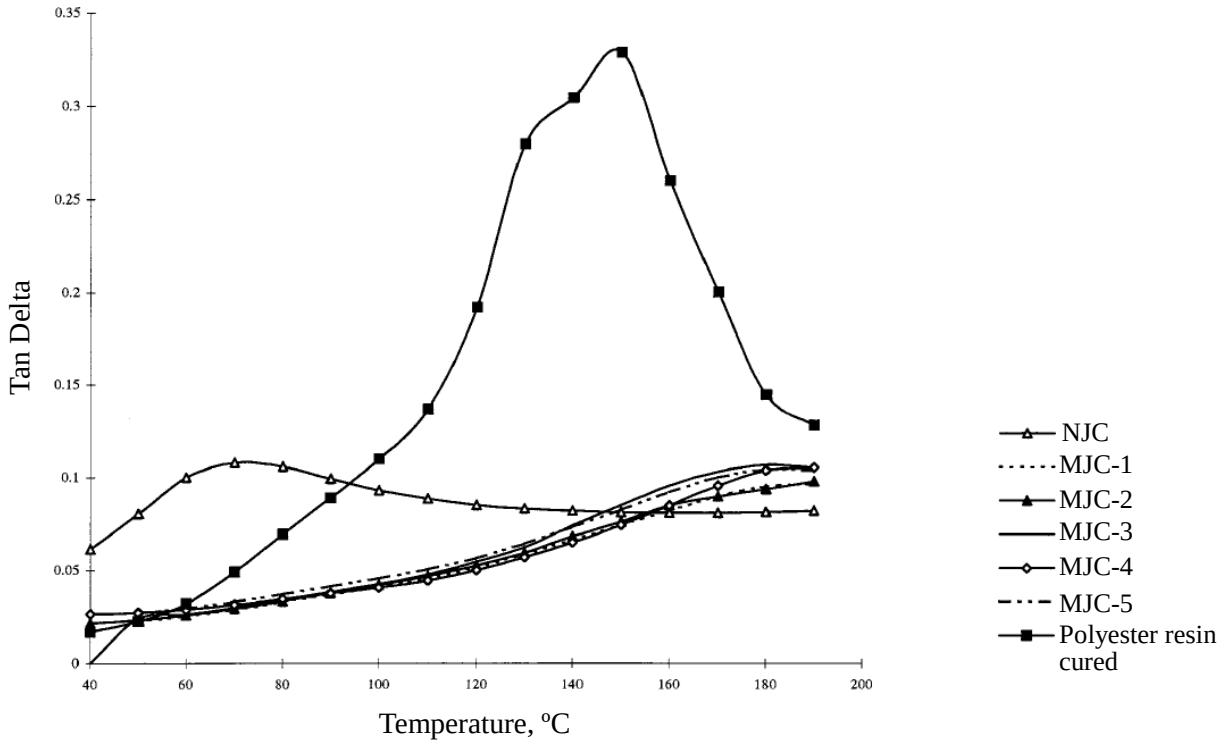

Figure 9. Variation in tan δ of neat polyester resin (cured), one unmodified jute-polyester composite (NJC), and five chemically modified (cyanoethylated) jute–polyester composites (MJC). Reprinted with permission from John Wiley and Sons [96].

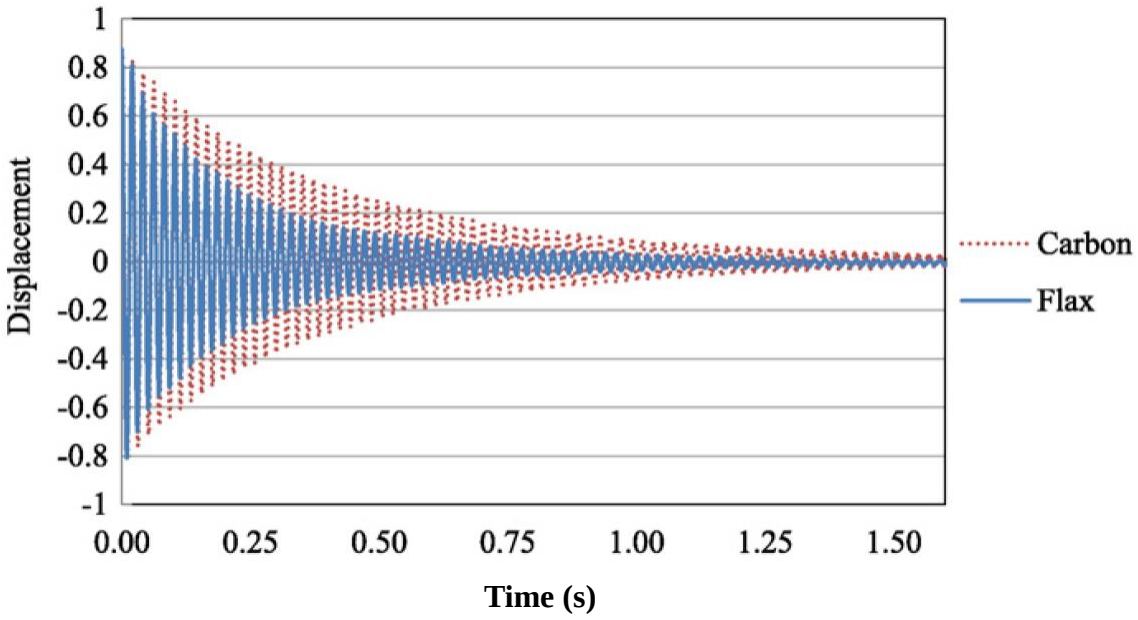

Figure 10. Vibration damping behaviors of carbon fiber and flax fiber reinforced composites. Reprinted with permission from Elsevier [107].

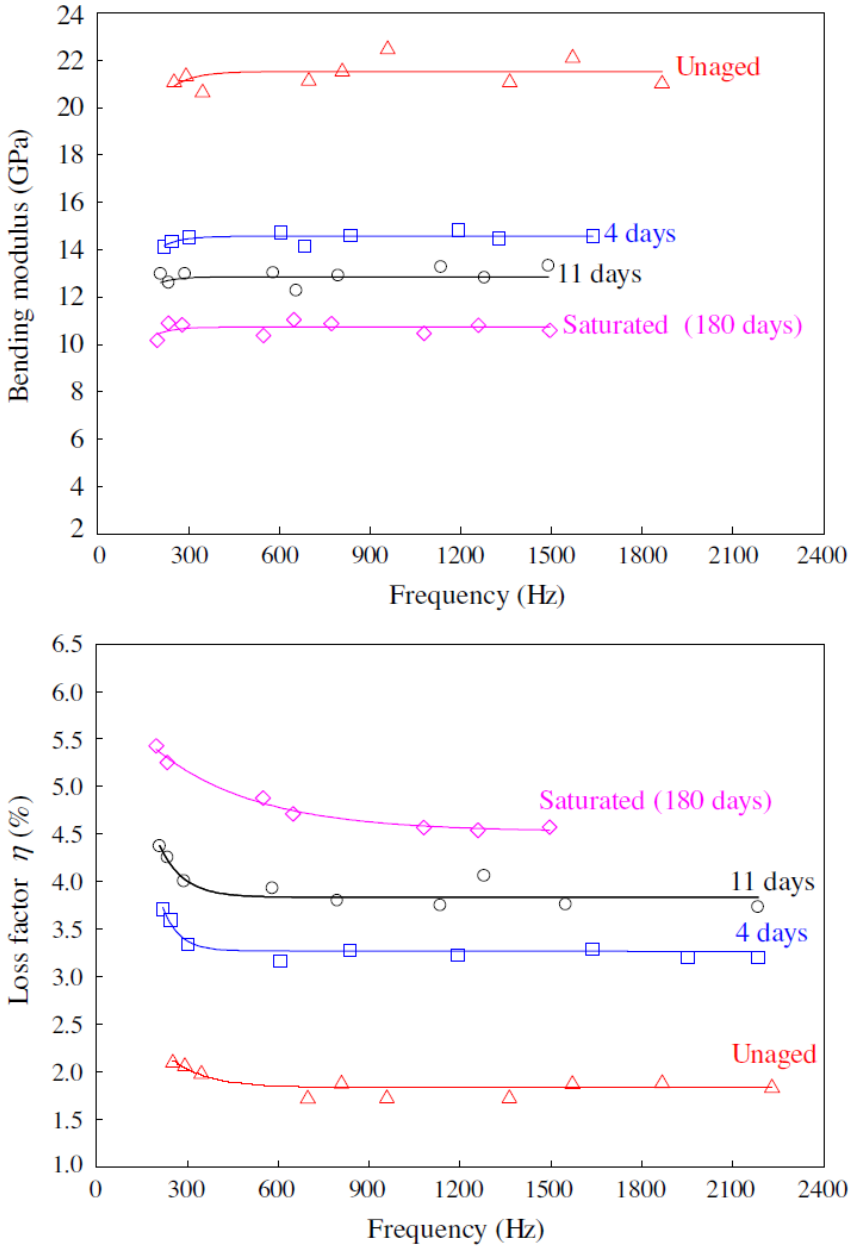

Figure 11. Variation in bending modulus and loss factor as a function of water aging time for flax-fiber reinforced epoxy composites. Reprinted with permission from Elsevier [110].